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^{137}Cs in freshwater fish and lake water in Finland after the Chernobyl deposition

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The uneven deposition of ^{137}Cs from the Chernobyl accident caused a large variation in the activity concentrations of ^{137}Cs in Finnish lakes. The concentrations of ^{137}Cs in lake water had decreased by more than 90% until the end of 1986, and thereafter the decrease gradually slowed down. In 2002, activity concentrations of ^{137}Cs in lake waters varied from 4 to 330 Bq m^{-3}. The concentrations of ^{137}Cs in fish had been generally highest in 1987 or 1988 and decreased thereafter by on average about 70% until 2005. After the deposition, several environmental processes, such as the water flow, sedimentation and runoff, and other characteristics such as potassium concentration, other water chemical parameters and the water residence time of the lake, have changed the distribution of ^{137}Cs in the compartments of the aquatic environment. The results also show that the activity concentrations of ^{137}Cs decreased at various rates in different lakes; ^{137}Cs in fish decreased three times more rapidly in one lake compared to another lake. In 2000–2004, activity concentration of ^{137}Cs in fish ranged from 8 Bq kg^{-1} to 7800 Bq kg^{-1} f.w. (f.w. = fresh weight).

Introduction

Deposition of ^{137}Cs from the Chernobyl accident was very unevenly distributed in Finnish lakes and their catchments. The fallout caused elevated activity concentrations of ^{137}Cs and there was a considerable variation both in lake waters and especially in freshwater fish. During the years since the deposition, ^{137}Cs has decreased in its concentration and redistributed in the components of the aquatic environment due to a variety of processes. Based on the distribution of the ^{137}Cs deposition in Finland, the country can be divided into five areas with various levels of ^{137}Cs deposition (Fig. 1) (Arvela *et al.* 1990). Besides the amount of ^{137}Cs deposited in the area, several lake and catchment specific environmental factors determine the long-term behaviour of ^{137}Cs in various lakes. The transport and retention of radiocesium in Scandinavian lakes during the first years after the deposition was eagerly studied (Andersson and Meili 1994, Björnstad *et al.* 1994, Notter *et al.* 1994, Saxén 1994). The large range in the activity concentrations of ^{137}Cs in fish and the existence of a large number of lakes of various types gave us the reason for the long-term study on behaviour of ^{137}Cs in the freshwater environment. The purpose of this study is to examine how the situation of ^{137}Cs in Finnish lakes has changed after 1986 and what the present situation is, twenty years after the fallout from Chernobyl.

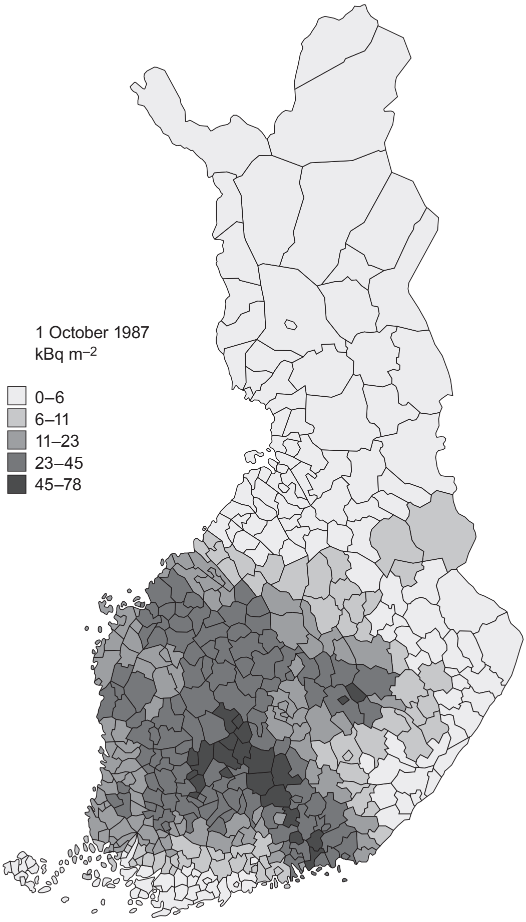

Fig. 1. Distribution of ^{137}Cs deposition in Finland and division of the country into five deposition areas (Arvela *et al.* 1990).

Material and methods

We analysed more than 7000 samples of freshwater fishes at the Radiation and Nuclear Safety Authority (STUK) after the Chernobyl deposition. About 400 lakes of various sizes and types were included in the study between 1986 and 2005. The sampling and analysing programme was at its largest in 1987 (Saxén 1990). Sampling was gradually focussed more and more on the most contaminated areas in Finland. Municipalities classified according to the number of the lakes sampled for ^{137}Cs in fish since 1986 are given in Fig. 2.

Lake water samples were analysed from frequent sampling of the southern part of lake Päijänne, the most important freshwater basin both in terms of freshwater fishing and as a source of drinking water for about one million inhabitants. The southern part of the lake is located in the area of the highest deposition in Finland. Additionally in 1998 and/or 2002, lake water was sampled and analysed from the same 30 or so lakes as the fishes were.

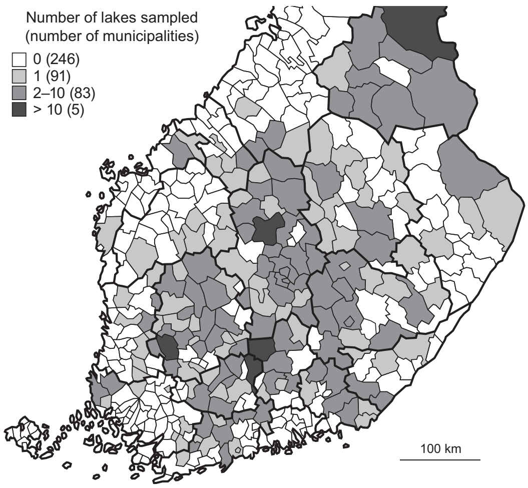

Fig. 2. Number of lakes sampled for fish in various municipalities in 1986–2005.

All the samples were analysed for gamma-emitting radionuclides using a gammaspectrometric method (Sinkko 1981, Klemola and Leppänen 1997).

Results

Lake water

In the waters of lake Päijänne, activity concentrations of ^{137}Cs decreased rapidly after the deposition in 1986 with an ecological half-time of 50 days. This means that the decrease was more than 90% already during 1986. Thereafter the decrease slowed down for a couple of years, the half-time being about one year at that time. Since 1989, the activity concentrations of ^{137}Cs decreased by half in about every five years (Fig. 3) (Saxén *et al*. 1996). The highest activity concentration of ^{137}Cs in the water of the southern part of lake Päijänne was 5600 Bq m^{-3} in May 1986 and in 2000 it was about 30 Bq m^{-3}. In lake water samples from about 30 lakes taken

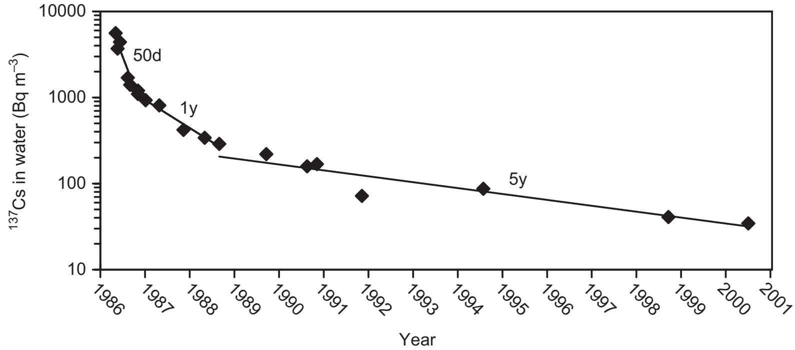

Fig. 3. Activity concentrations of ^{137}Cs in water of the southern part of lake Päijänne with ecological halftimes of ^{137}Cs.

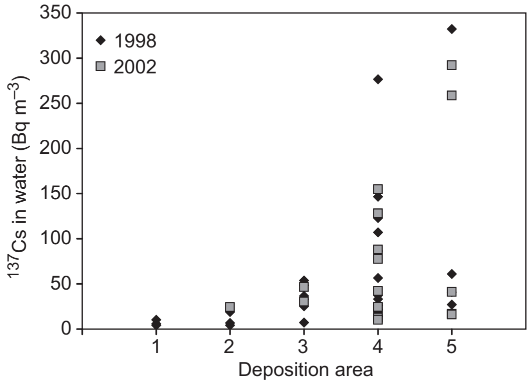

Fig. 4. Activity concentrations of ^{137}Cs in lake waters in 1998 and 2002 in various deposition areas given in Fig. 1.

in 1998 and/or 2002, activity concentrations of ^{137}Cs varied from 4 to 330 Bq m^{-3} (Saxén 2005). The whole range of values was recorded in the deposition areas four and five (Fig. 4). Compared with the original deposition on the sampling municipality corrected for radioactive decay, the water concentrations still seemed to correlate relatively well with the deposition in certain lakes, while in others the activity concentrations were much lower than could be expected on the basis of the deposition (Fig. 5).

Fish

Fishes get ^{137}Cs from their food, therefore the non-predatory fish reached their maximum ^{137}Cs contents already in the same year as the deposition occurred, while the predatory fish like pike and large perch had their maximum a year or two later due to the longer food chain. Activity concentrations of ^{137}Cs in predatory fish had peak values in 1987 and in some cases in 1988. The range of ^{137}Cs concentrations in fish was large throughout the study period, especially in the deposition area five (Fig. 6). In 2000–2004, the activity concentrations of ^{137}Cs in fish varied

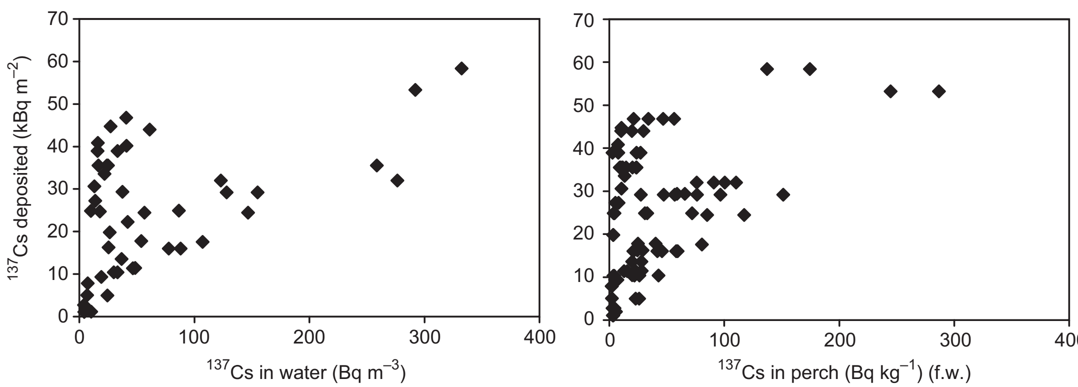

Fig. 5. ^{137}Cs in lake water and in perch (*Perca fluviatilis*) versus ^{137}Cs deposited in the sampling municipality in 1998 and 2002. The values for the deposited ^{137}Cs were corrected for radioactive decay to the sampling date of water and fish.

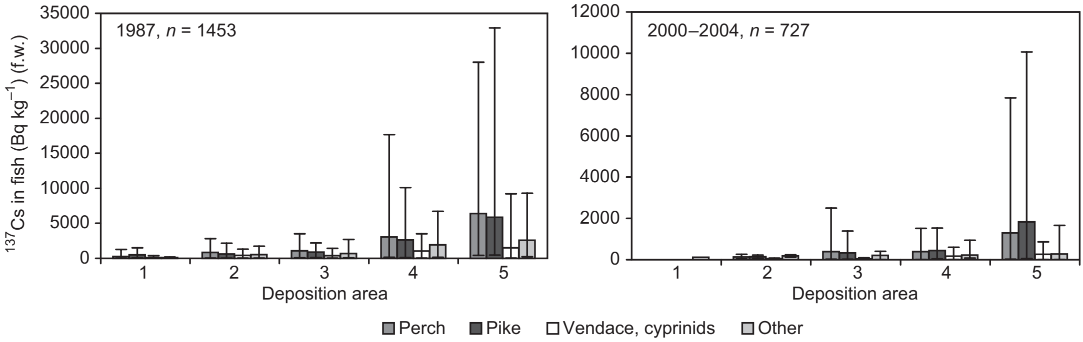

Fig. 6. Average activity concentrations of ^{137}Cs and variation ranges in various species of fish in different deposition areas in 1987 and 2000–2004 (for deposition areas *see* Fig. 1).

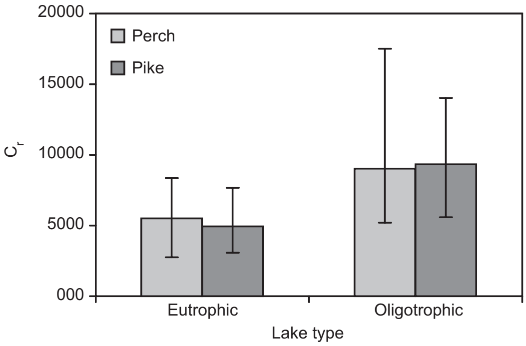

Fig. 7. Average concentration ratios (C_r = ^{137}Cs in fish/^{137}Cs in water) with variation in oligotrophic and eutrophic lakes (five lakes are included in both lake types).

from 8 to 7800 Bq kg^{-1} (f.w.). The activity concentrations decreased on average by about 70% from 1987 to 2000–2004. Within a lake the intra-specific variation in ^{137}Cs concentrations was a factor of 2–3, while the corresponding inter-specific variation reached a factor of 2–10 in 2000–2004.

Transfer of ^{137}Cs into fish is commonly described by means of the concentration ratio, C_r, defined as C_r = (^{137}Cs Bq kg^{-1} (f.w.) in fish)/(^{137}Cs Bq kg^{-1} in water)). In our study the values of C_r in oligotrophic lakes were on average two and in some cases even ten times higher than those in eutrophic lakes (Fig. 7) (Saxén 2006). The values of C_r varied by a factor of 20 in all the lakes studied.

The results indicate further that the concentrations of ^{137}Cs in fish decrease at various rates in different lakes. Examples of this are given in Fig. 8 which shows that in pike (*Esox lucius*) from lake Pälkänevesi activity concentration of ^{137}Cs decreased more than twice as quickly as that in pike from lake Pieni Ahveninen. This is described by ecological half-times which for ^{137}Cs in pike were about four years and nine years, respectively, in these two lakes in 1988–2002. The largest difference in the ecological halftimes of ^{137}Cs in fish from various lakes that we noticed in our study was from three to nine years (Saxén 2006). The decrease rates of the

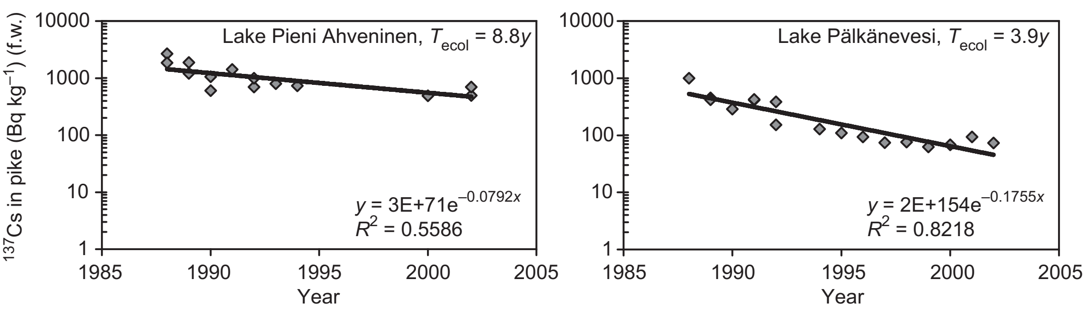

Fig. 8. Decrease of activity concentrations of ^{137}Cs in pike (*Esox lucius*) in two lakes (Pieni Ahveninen and Pälkänevesi) with the observed ecological halftimes.

^{137}Cs concentrations in pike and perch (*Perca fluviatilis*) from the same lake were almost the same. This argues that differences between lakes are much higher than differences between species.

Although our fish sampling in 2000–2004 was focussed on the areas of the highest deposition, the frequency distribution of all activity concentrations in those years was close to log-normal distribution. The mean and median activity concentrations of ^{137}Cs in fish were 230 and 200 Bq kg^{-1}, respectively, in 2000–2004. In the Finnish catch of freshwater fish as a whole, the activity concentration of ^{137}Cs would be lower because almost half of the catch comes from eastern Finland with relatively low deposition.

Discussion

The activity concentration of ^{137}Cs in lake waters decreased quickly after the deposition and is now at a relatively low level, even though the concentrations vary by a factor of about one hundred. The effects of various environmental processes, such as the retention of ^{137}Cs to particles and sinking onto the bottom sediment, resuspension of ^{137}Cs from the sediment back to the water, as well as amount of water exchange and runoff, are reflected in the changes of the decrease rate, described by the observed ecological halftimes of ^{137}Cs in water, at various times. The radiation dose caused by ^{137}Cs in drinking water is at the moment only of the order of one microsievert per year or less, even if one would use lake water with the highest observed activity concentrations of ^{137}Cs as drinking water.

According to a Swedish study (Sundbom *et al*. 2003), the peak concentrations of ^{137}Cs in different fish were attained 56–806 days after the fallout. This delay increased with the increasing body size and trophic level of the fish, as seen also in this study.

Earlier post-Chernobyl studies have already shown that inter-lake differences in the transfer of ^{137}Cs from fallout into fish could not be explained by the difference in the growth rate of fish (pike) between various lakes. The mean hydraulic residence time and the scavenging capacity of the lakes seemed to affect the total transfer of ^{137}Cs into fish (Andersson and Meili 1994). Radionuclides that deposited directly onto the water system form the primary load of a lake. The binding of cesium to the particulate material of the water and its sinking with this material onto the bottom sediment lower the availability of the ^{137}Cs for fish. This process is influenced by the sedimentation rate and the amount of suspended solids in the lake. A long water residence time and slow flow rate maintain the cesium available for food chains in a lake.

Kolehmainen *et al*. (1966) documented that the activity concentration of ^{137}Cs in fish was inversely proportional to the potassium content of the water. In accordance with this, in our study the uptake of ^{137}Cs by fish was affected by the nutrition level of the lake water, being higher in the oligotrophic lakes as compared with the eutrophic ones.

The amount of the secondary load from the catchment to the lake, affected e.g. by soil type, land use, topography and other characteristics of the catchment, explains partly the inter-lake differences in the ecological half-times of ^{137}Cs observed in fish, as do also differences in water residence times and flow rates. Different rates of decrease in the radionuclide levels in various water bodies were also noticed in the lakes near the Chernobyl nuclear power plant. The rate of fish decontamination was noticed to be connected with the hydrology and hydrochemistry of the water body in addition to the initial amount of the deposited radionuclides. The main reasons for the high activity concentration of ^{137}Cs in water, sediment and fish were discovered to be low content of K and very slow water exchange (Ryabov 2002). Ultimately, the joint effect of a large variety of factors affecting the levels of radionuclides in water, the uptake of ^{137}Cs by fish or the availability of ^{137}Cs in the food chain, finally determines the fate of ^{137}Cs in a lake and explains the differences in the rates of the decline of the radionuclide levels in fish. The fish data obtained in our study were utilized for developing a statistical model that predicts ^{137}Cs activity concentration levels in fish regionally (Saxén and Sundell 2006). However, due to the large number of simultaneously affecting factors, it is difficult to predict exactly the levels of ^{137}Cs in fish in a lake.

Conclusions

Twenty years after the Chernobyl accident, the activity concentration of ^{137}Cs in lake waters is at a low level, even though the concentrations vary by a factor of about one hundred (4–300 Bq m^{-3}). In many lakes, especially in northern and eastern Finland, the activity concentration of ^{137}Cs in fish has decreased to the pre-Chernobyl levels many years ago. These levels were in the range 2–64 Bq kg^{-1} (f.w.) in fish sampled from certain important lakes for fishery in 1982 (STL 1983). In Finland, the pre-Chernobyl level values are presently found also in the areas of the highest Chernobyl deposition. However in the areas of the highest deposition in Finland, in some lakes characterised by a high uptake of ^{137}Cs by fish and/or a slow decline of ^{137}Cs in fish and/or a long water residence time, low sedimentation rate and/or high transfer of ^{137}Cs from the catchment to the lake, predatory fish may still have activity concentrations of several thousands Bq kg^{-1} (f.w.). In these lakes it will take several years, even decades, before activity concentrations of ^{137}Cs in predatory fish decrease to the pre-Chernobyl level.

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